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Model Simulations of the First Aerosol Indirect Effect and Comparison of Cloud Susceptibility to Satellite Measurements

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INTRODUCTION

Present-day global anthropogenic emissions contribute more than half of the mass in sub-micron particles primarily due to sulfate and carbonaceous aerosol components derived from fossil fuel combustion and biomass burning. These anthropogenic aerosols modify the microphysics of clouds by serving as cloud condensation nuclei (CCN) and enhance the reflectivity of low-level water clouds, leading to a cooling effect on climate (the Twomey effect or first indirect effect). The magnitude of the first aerosol indirect effect is associated with cloud frequency as well as a quantity representing the sensitivity of cloud albedo to changes in cloud drop number concentration. This quantity is referred to as cloud susceptibility [*Twomey*, 1991]. Analysis of satellite measurements demonstrates that marine stratus clouds are likely to be of higher susceptibility than continental clouds because of their lower number concentrations of cloud drops [*Platnick and Twomey*, 1994]. Here, we use an improved version of the fully coupled climate/chemistry model [Chuang et al., 1997] to calculate the global concentrations of sulfate, dust, sea salt, and carbonaceous aerosols (biomass smoke and fossil fuel organic matter and black carbon). We investigated the impact of anthropogenic aerosols on cloud susceptibility and calculated the associated changes of shortwave radiative fluxes at the top of the atmosphere. We also examined the correspondence between the model simulation of cloud susceptibility and that inferred from satellite measurements to test whether our simulated aerosol concentrations and aerosol/cloud interactions give a faithful representation of these features.

CLOUD SUSCEPTIBILITY

A cloud drop parameterization has been applied in the model to calculate the concentrations of cloud drops (N_d) nucleated on different aerosol components. This parameterization was based on the mechanism of drop formation and the chemical processes controlling the formation of anthropogenic sulfate [Chuang et al., 1997]. It is assumed that anthropogenic sulfate is deposited on pre-existing particles that are externally mixed with natural sulfate, dust, sea salt, and carbonaceous aerosols. The addition of anthropogenic sulfate onto pre-existing particles does not change the total aerosol number, but the resulting particle distribution grows to larger sizes. We found that the increases of N_d can be up to 200 cm^{-3} due to anthropogenic carbonaceous aerosols and up to 30 – 90 cm^{-3} due to the change of aerosol size distribution associated with anthropogenic sulfate. Our simulations indicate that the presence of industrial aerosols has significantly reduced cloud susceptibility in the northern hemisphere in January, and biomass aerosols have reduced susceptibility in the southern hemisphere in July.

Figure 1 presents the distributions of model simulated susceptibility for warm clouds and those derived from satellite-retrieved values of cloud optical thickness τ and effective drop radius r_e. These data were retrieved from 1989 - 1991 AVHRR measurements using the algorithm developed by Kawamoto et al. [2000]. The column cloud drop number concentration $N_{d,col}$ was estimated from the retrieved τ and r_e under assumptions of a vertically homogeneous drop profile and a log-

normal drop size distribution with a standard deviation $\sigma = 1.4$. To obtain N_d from the satellite-retrieved $N_{d,col}$, a value of liquid water content 0.30 g m^{-3} is used here to estimate the cloud thickness from the liquid water path. As shown in Figure 1, the general features of the derived cloud susceptibility are similar to those of the model, though the magnitude is higher by about a factor of 2 in most of the regions. This discrepancy may be caused by the uncertainty in the prescribed drop size distribution where the retrieved $N_{d,col}$ varies with the assumed magnitude of σ. For $r_e = 10$ μm and $\tau = 6$, the retrieved $N_{d,col}$ would be 34% lower if σ decreases from 1.4 to 1.1 and would be 288% higher if $\sigma = 2.0$. There are further uncertainties associated with the prescribed liquid water content and the retrieved cloud top temperature. An increase in liquid water content from 0.30 g m^{-3} to 0.35 g m^{-3} would reduce the derived susceptibility by 17%. A temperature error of 2.5°K could cause an error in retrieved r_e of about 7% [Kawamoto et al., 2000] which would lead to a comparable uncertainty in susceptibility. Moreover, the uncertainty of the model simulation itself may also contribute to the discrepancy.

FIRST INDIRECT FORCING BY ANTHROPOGENIC AEROSOLS

Figure 2a presents the simulated first indirect forcing by anthropogenic carbonaceous aerosols for January and July. In general, the forcing in July is stronger than that in January, and yields a global average of –1.59 W m^{-2} and –1.05 W m^{-2}, respectively. The maximum value is about –7.7 W m^{-2} along the west coast of Mexico in January and –8.6 W m^{-2} along the east coast of Brazil in July. Anthropogenic carbonaceous aerosols together with natural particles are treated as an external mixture in the cloud drop parameterization and lead to an annual average forcing of –1.51 W m^{-2}. This value is much lower than our previous study (–2.5 to –4.5 W m^{-2}, see Penner et al., 1996) in which part of natural emissions were absent. Figure 2b shows the first indirect forcing by anthropogenic sulfate deposited onto pre-existing particles derived from natural emissions and anthropogenic carbonaceous sources. The maximum forcing is about –1.6 W m^{-2} in January and –5.1 W m^{-2} along the east coast of the United States. While the forcing pattern is similar to that calculated previously in Chuang et al. [1997], the forcing magnitude is considerably smaller and leads to a global annual average of –0.30 W m^{-2}. Current calculations used the model-generated aqueous sulfate production rate. This rate is approximately 88% of the total sulfate source strength. These simulations may, therefore, be compared to the previously calculated forcing of –0.41 W m^{-2} for a case of prescribed 85% sulfate production through the aqueous pathway [Chuang et al., 1997]. The present values are smaller both because of the larger source strength for natural organic aerosols and because of the presence of sea salt and dust. Figure 3 indicates that the global average of the first indirect forcing by total anthropogenic aerosols is largest in April-June associated with tropical biomass burning of savanna and forested areas. Its magnitude varies seasonally from –1.2 W m^{-2} in January to –2.4 W m^{-2} in May and yields a global annual average of –1.85 W m^{-2}. Because of the nonlinear relationship between cloud drop number and aerosol number concentrations, the total forcing does not equal the sum of the forcing from each individual source.

SUMMARY

Our simulations are consistent with the analysis of satellite retrieved cloud susceptibility and demonstrate that marine stratus clouds are more sensitive to changes in cloud drop number concentration than continental clouds. We note that our cloud drop parameterization does not include the impact of drop spectral broadening on the cloud susceptibility. The dependence of cloud susceptibility on drop number concentration is larger for clouds with an active collection process [Feingold et al., 1997]. In spite of this shortcoming in the cloud drop parameterization, our simulations provide a global understanding of the effects of anthropogenic aerosols on water cloud susceptibility and global radiation budget.

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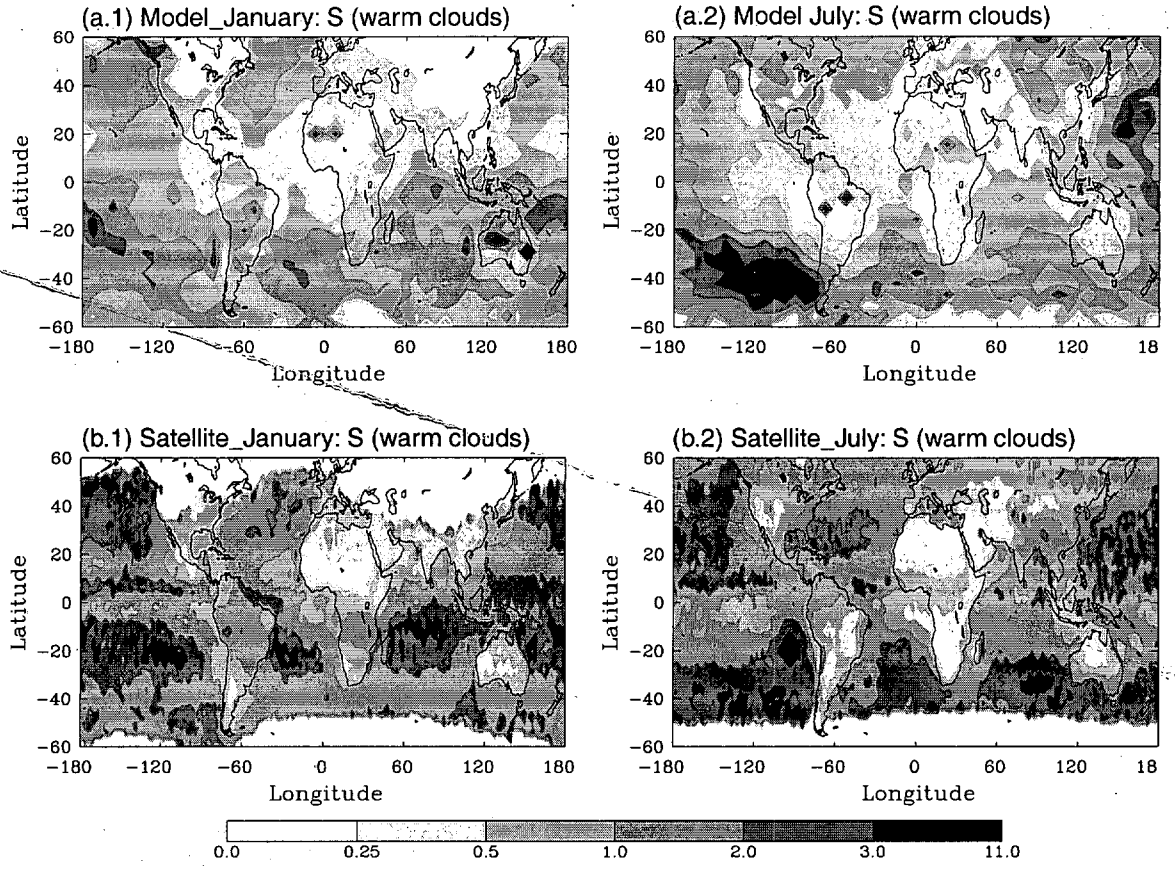

Fig. 1. Comparison of model calculated cloud susceptibility ($\times 10^{-3}$ cm^3) for warm clouds with those inferred from satellite measurements.

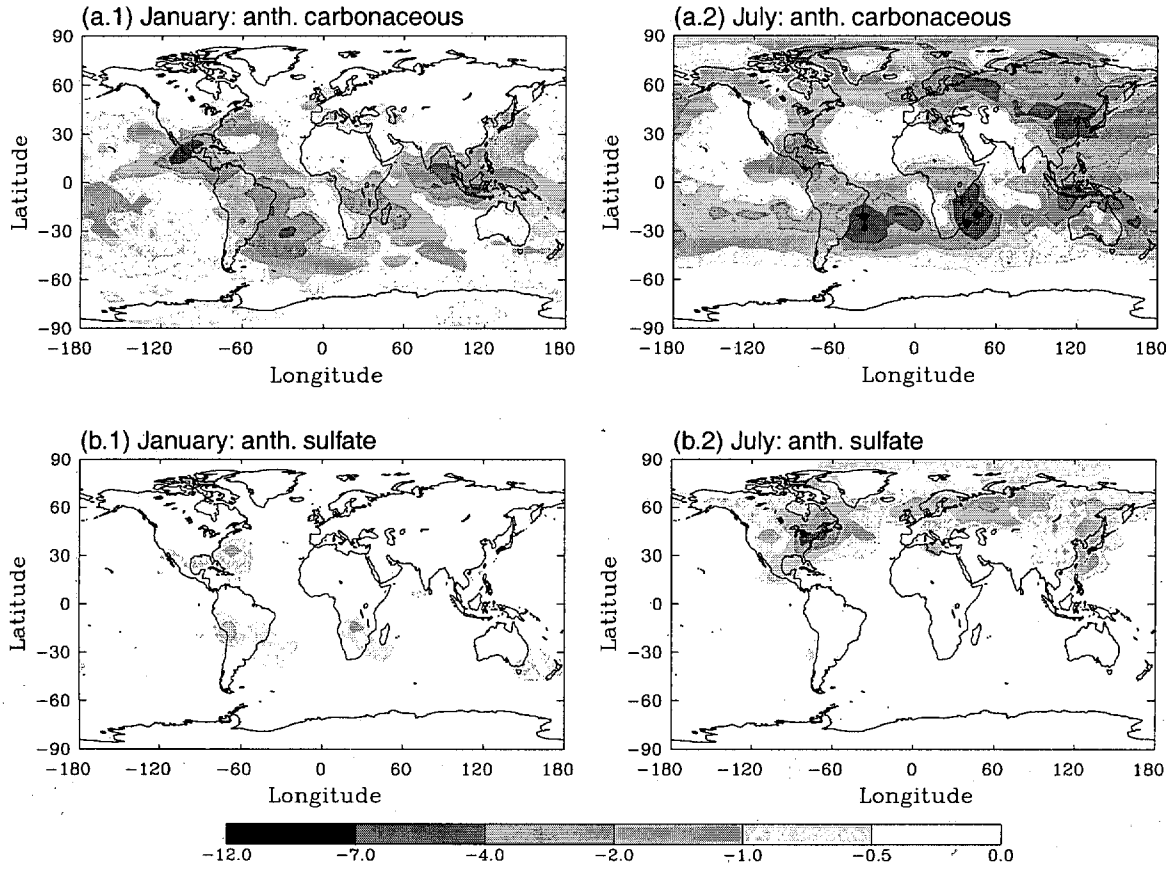

Figure 2. Simulated monthly average aerosol indirect forcing (W m^{-2}) due to (a) externally mixed anthropogenic carbonaceous aerosols and (b) deposition of anthropogenic sulfate onto pre-existing particles (natural particles + anthropogenic carbonaceous particles).

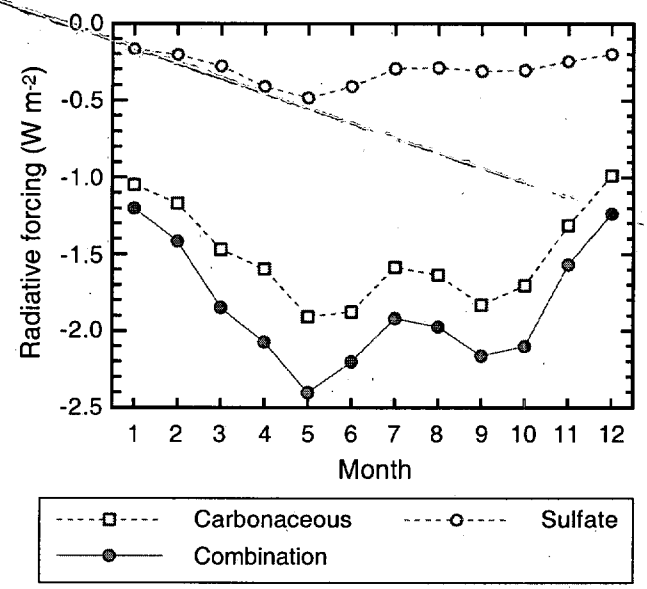

Figure 3. Seasonal variations of simulated global average of the first indirect forcing (W m^{-2}).